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(54) **SILICE PRECIPITE**
(54) **PRECIPITATED SILICA**

$$\frac{d_{90} - d_{10}}{2d_{50}}$$

(57) Precipitated silica which has the following physico-chemical parameters: BET surface area (DIN 66131) in m^2/g 400 - 600, DBP index (DIN 53601) in g/100 g 300 - 360, Compacted density (DIN 53194) in g/l 70 - 140, Grindometer value (ISO 1524) in µm 15 - 50, Size distribution index I < 1.0, measured with a Malvern instrument, Size distribution index I = (see above formula). It is prepared by milling a precipitated silica in a classifier mill or a fluidised bed counter flow mill. A polyethylene wax emulsion may be added before the milling procedure. The precipitated silica then has the following physico-chemical parameters: BET surface area (DIN 66131) in m^2/g 351 - 600, DBP index (DIN 53601) as a % 300 - 360, Carbon content as a % 1 - 8, Compacted density (DIN 53194) in g/l 70 - 140, Grindometer value (ISO 1524) in µm 15 - 50, Size distribution index I < 1.0. The precipitated silicas may be used as matting agents in lacquer systems.

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ABSTRACT

Precipitated silica which has the following physico-chemical parameters:

BET surface area (DIN 66131) in m^2/g	400 - 600
DBP index (DIN 53601) in g/100 g	300 - 360
Compacted density (DIN 53194) in g/l	70 - 140
Grindometer value (ISO 1524) in µm	15 - 50
Size distribution index I	< 1.0

measured with a Malvern intrument

$$\text{Size distribution index I} = \frac{d_{90}-d_{10}}{2d_{50}}$$

It is prepared by milling a precipitated silica in a classifier mill or a fluidised bed counter-flow mill. A polyethylene wax emulsion may be added before the milling procedure. The precipitated silica then has the following physico-chemical parameters:

BET surface area (DIN 66131) in m^2/g	351 - 600
DBP index (DIN 53601) as a %	300 - 360
Carbon content as a %	1 - 8
Compacted density (DIN 53194) in g/l	70 - 140
Grindometer value (ISO 1524) in µm	15 - 50
Size distribution index I	< 1.0

The precipitated silicas may be used as matting agents in lacquer systems.

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The embodiments of the invention in which an exclusive property or privilege is claimed are defined as follows:

1. Precipitated silica having the following physico-chemical parameters:

BET surface area (DIN 66131) in m^2/g	400 - 600
DBP index (DIN 53601) in g/100 g	300 - 360
Compacted density (DIN 53194) in g/l	70 - 140
Grindometer value (ISO 1524) in µm	15 - 50
Size distribution index I measured with a Malvern intrument	< 1.0

$$\text{Size distribution index } I = \frac{d_{90} - d_{10}}{2d_{50}}$$

2. A process for preparing precipitated silica with the physico-chemical parameters as defined in claim 1, in which a precipitated silica which has the following physico-chemical characteristics:

BET surface area (DIN 66131) in m^2/g	400 - 600
DBP index (DIN 53601) as a %	340 - 380
Compacted density (DIN 53194) in g/l	180 - 220
"Alpine" sieve residue > 63 µm wt.%	25 - 60,

is milled using a classifier mill or a fluidised bed counter-flow mill.

3. Precipitated silica coated with a polyethylene wax emulsion, having the following physico-chemical parameters:

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BET surface area (DIN 66131) in m^2/g	351 - 600
DBP index (DIN 53601) as a %	300 - 360
Carbon content as a %	1 - 8
Compacted density (DIN 53194) in g/l	70 - 140
Grindometer value (ISO 1524) in µm	15 - 50
Size distribution index I	< 1.0.

4. A process for preparing precipitated silica coated with polyethylene wax emulsion as defined in claim 3, in which a polyethylene wax emulsion is added to a precipitated silica which has the following physico-chemical characteristics:

BET surface area (DIN 66131) in m^2/g	400 - 600
DBP index (DIN 53601) as a %	340 - 380
Compacted density (DIN 53194) in g/l	180 - 220
"Alpine" sieve residue > 63 µm wt.%	25 - 60,

and the mixture is then dried and milled using a classifier mill or a fluidised bed counter-flow mill.

5. A process according to claim 4, in which the precipitated silica is prepared, a resultant filter cake is liquefied under the action of shear forces, polyethylene wax emulsion is added, and the mixture is spray dried and milled using a classifier mill or a fluidised bed counter-flow mill.

6. Use of precipitated silica in accordance with claim 1 or 3 as a matting agent in lacquer systems.

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Precipitated Silica

The invention relates to precipitated silica, a process for its preparation, and its use as a matting agent.

It is known that synthetic, precipitated silicas or silica gels can be used as matting agents (DE-PS 24 14 478, DE-PS 17 67 332, DE-OS 16 69 123, DE-AS 15 92 865, DE-A 38 15 670).

The matting power of a silica depends on a variety of factors, such as, for example, the type of silica, the particle size, the particle size distribution, the refractive index and also the lacquer system. The shape and size distribution of secondary particles in the silica are of particular importance.

In addition to being very efficient, expressed by the reduction in degree of gloss as compared with the non-matted lacquer film, a silica which is used as a matting agent also has to satisfy a number of other requirements. Thus, for example, there should be no undue thickening of the lacquer system due to the silica which is introduced. A smooth surface to the lacquer should be produced on the corresponding thin lacquer coatings. Specks which have an adverse effect on the surface quality must be avoided.

The document DE-A 31 44 299 describes precipitated silicas and a process for preparing these precipitated silicas,

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which are characterised by the following physico-chemical properties:

BET surface area according to DIN 66131 in m^2/g	400 - 600
DBP index according to DIN 53601 as a %	320 - 360
and	
BET surface area according to DIN 66131 in m^2/g	400 - 600
DBP index according to DIN 53601 as a %	310 - 360
Compacted density according to DIN 53194 in g/l	75 - 120
"Alpine" sieve residue > 63 µm in wt.%	< 0.1

When preparing these silicas, an Alpine transverse flow mill or a jet mill is used to mill the product following spray drying. It is also specified in this document that these precipitated silicas are valuable, highly effective matting agents for lacquers. Precipitated silicas which are prepared using these types of mills lead to disadvantageous roughness of the surface due to the presence of large specks in the final lacquer. The grindometer value (according to ISO 1524) in black stoving enamel is greater than 100 µm and 85 to 90 µm respectively for the known precipitated silicas. Thus these precipitated silicas can only be used to a limited extent as matting agents.

It is an object of this invention to provided a precipitated silica which minimises these disadvantages.

Precipitated silca according to this invention is characterised by the following physico-chemical parameters:

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BET surface area according to DIN 66131 in m^2/g	400 - 600
DBP index according to DIN 53601 in g/100 g	300 - 360
Compacted density according to DIN 53194 in g/l	70 - 140
Grindometer value according to ISO 1524 in µm	15 - 50
Size distribution index I measured with a Malvern instrument	< 1.0

Size distribution index $I = \frac{d_{90}-d_{10}}{2d_{50}}$

Another aspect of the invention provides a process for preparing the precipitated silicas according to the invention in which a precipitated silica which has the following physico-chemical properties:

BET surface area according to DIN 66131 in m^2/g	400 - 600
DBP index according to DIN 53601 as a %	340 - 380
Compacted density according to DIN 53194 in g/l	180 - 220
"Alpine" sieve residue > 63 µm wt.%	25 - 60,

is milled using a classifier mill or a fluidised bed counter-flow mill.

The initial silica is described in the document DE-A 31 44 299.

By way of example, a ZPS classifier mill (Zirkoplex® Alpine Aktiengesellschaft D-8900 Augsburg), or an AFG fluidised bed counter-flow mill may be used.

In one embodiment of the invention, the precipitated silica according to the invention may be classified after milling, in order to adjust to a specific granular fraction.

Classifying may be performed, for example, using an ATP Turboplex fine classifier (Alpine Aktiengesellschaft D-8900 Augsburg).

Another aspect of the invention provides a precipitated silica coated with a polyethylene wax emulsion, which is characterised by the following physico-chemical parameters:

BET surface area according to DIN 66131 in m^2/g	351 - 600
DBP index according to DIN 53601 as a %	300 - 360
Carbon content as a %	1 - 8
Compacted density according to DIN 53194 in g/l	7 - 140
Grindometer value according to ISO 1524 in µm	15 - 50
Size distribution index I	< 1.0

This precipitated silica can be prepared by adding polyethylene wax emulsion to a precipitated silica which has the following physico-chemical characteristics:

BET surface area according to DIN 66131 in m^2/g	400 - 600
DBP index according to DIN 53601 as a %	340 - 380
Compacted density according to DIN 53194 in g/l	180 - 220
"Alpine" sieve residue > 63 µm wt.%	25 - 60,

and then drying and milling the product using a classifier mill or a fluidised bed counter-flow mill.

In a particular embodiment of the invention, the precipitated silica can be prepared by liquefying filter cake under the action of shear forces, adding polyethylene wax emulsion, spray drying, and then milling using a classifier mill or a fluidised bed counter-flow mill.

A precipitated silica in accordance with DE-A 31 44 299 is preferably used as the starting silica.

An advantage of precipitated silicas according to the invention is in particular their high matting efficiency, in addition to further advantages such as providing a very smooth surface of the dry lacquer, high transparency and a small effect on the rheology (viscosity) of the lacquer.

The invention will be further described and exemplified in the following description, which makes reference to the accompanying drawings, in which:

Figure 1 shows the size distribution of classified precipitated silica.

Figure 2 shows the particle size distribution of precipitated silicas according to the invention, compared to the particle size distribution of a precipitated silica in accordance with DE-A 31 44 299.

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Examples

Example 1

A precipitated silica prepared in accordance with example 1 from DE-A 31 44 299 is milled in a ZPS 100 Zirkoplex® classifier mill from the Alpine company, by varying the throughput and the process parameters such as speed of rotation of the classifier, milling throughput and milling air. The trial parameters, the physico-chemical data and the paint properties which are obtained in black stoving lacquer are given in table 1.

Example 2

A precipitated silica prepared in accordance with example 1 from DE-A 31 44 299 is milled in an AFG 200/1 fluidised bed counter-flow mill, from the Alpine company, while varying the throughput and the process parameters such as rate of rotation of the classifier, or the milling air. The trial parameters, the physico-chemical data and the paint properties which are obtained in black stoving lacquer are given in table 2.

Example 3

Precipitated silicas which are prepared in accordance with example 1c or example 2c (see table 1 and 2) are classified in an ATP 50 turboflex fine classifier to give a finer and a coarser fraction. The process parameters, the physical data and the paint test results which are obtained in black stoving lacquer are given in table 3.

Example 4 (comparison example)

The unmilled, spray-dried silica, prepared in accordance with DE 31 44 299 (example 6), is milled on a UP 630 Alpine

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transverse flow mill. The physico-chemical data and paint properties of the product obtained are given in table 4.

Example 5 (comparison example)

The unmilled, spray-dried silica, prepared in accordance with DE 31 44 299 (example 9), is milled using an MC 500 Microgrinding air jet mill. The physico-chemical data and paint properties are given in table 4.

The effectiveness and matting efficiency of the precipitated silicas prepared according to examples 1 to 3 are tested in a black stoving lacquer. The Lange gloss values, at angles of reflection of 60° and 85°, and the Hegman grindometer value were also assessed.

The B. Lange gloss meter was used to determine the degree of gloss, which is a measure of the matting power of the matting silica tested. The B. Lange gloss meter uses angles of incidence and reflection of 60° and 85°. The degrees of gloss measured are cited as percentages. The lower this value, the better is the matting capacity of the precipitated silica. As a result, less matting agent has to be used in order to achieve a quite specific degree of gloss or a specified matting effect.

The grindometer value is determined using a grindometer. The grindometer value, which is measured in µm (micrometers) is a measure of the largest particles which can be found after stirring the precipitated silica into the final, sprayable lacquer mixture. It can be related to the production of specks in the dry lacquer film, so undesired specks or sprayed granules can be detected using the grindometer (ISO 1524).

The quality of the lacquer film surface is determined using the scanning section method developed by the Hommelwerke

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company and is cited as an average roughness value (Ra) according to DIN 4768/1, DIN 4762/1E and as an average depth of roughness (RZD) according to DIN 4768/1.

The black stoving lacquer used had the following composition:

	Parts by wt
Carbon black paste, tack 1	8.0
Jägalyd R40, 60 % strength in xylene	50.8
Maprenal MF 800, 55 % strength in butanol	25.9
Baysilone paint additive OL 17,1 % in xylene	2.0
Thinner	13.3
	100.0

Thinner	
Xylene	75.0
Butanol	10.0
Ethoxypropanol	15.0
	100.0

4 g of precipitated silica are stirred into 100 g of lacquer with a blade stirrer at 2000 rpm for 10 minutes. The viscosity of the mixture is adjusted to a flow time of 20 seconds using xylene (DIN; 4 mm nozzle).

The lacquer is sprayed to give an approximately 30 µm thick dry layer on sheet metal, air dried and fired at 180°C for 30 minutes.

Example 6

The paint properties of the precipitated silicas prepared according to examples 1a to c, a precipitated silica prepared according to DE 38 15 670 and a commercially available product (Nipsil 1009) are tested in two other test lacquer systems.

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CC lacquer

	Parts by wt.
Alftalat AN 950, 60% in Solvesso 150/Butylglycol	29.30
Solvesso 150	2.60
Titanium dioxide Kronos 2059	33.60
Aerosil R 972	0.20
Dispersion: 40 h ball mill KU 5, 60 rpm, 4900 g Alubite beads 19 mm	
Alftalat AN 950, 60 % in Solvesso 150/Butyl glycol	13.00
Maprenal MF 900, 100 %	8.10
Maprenal MF 577, 50 % in butanol	0.80
Butyl glycol	2.00
Solvesso 150	2.90
Xylene	6.70
DOW CORNING PA 57	0.60
p-Toluylsulfonic acid, 20 % in butanol	0.30
Total	100.00

Before use, 3.2 g of matting agent are dispersed in 150 parts by weight of lacquer using a blade stirrer at 2000 rpm.

DD lacquer

	Parts by wt.
CAB 381-0,5	0.3
Butyl acetate, 98 % strength	11.0
Ethoxypropyl acetate	16.5
Desmophen 800	15.0
Desmophen 1100	20.0
Mowilit, 50 % strength in ethyl acetate	3.0
Baysilone-lacquer additive	0.1
Xylene	34.1
Total	100.00

Firstly 0.3 parts by weight of CAB 381-0.5 are carefully dissolved in 11.0 parts by weight of butyl acetate (98.0 % strength) and 16.5 parts by weight of ethoxypropyl acetate using a high speed stirrer. Then the other components are

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added in the sequence given above and the mixture is homogenised by stirring.

Before use, the gloss lacquer is homogenised with the blade stirrer. The matting agent (amount see table 6) is dispersed in 100 parts by weight of lacquer using a blade stirrer at 2000 rpm. After a degassing time of 15 minutes, 50 g of the hardener Desmodur L 75 are added and homogenised with the blade stirrer for 2 minutes at 1000 rpm. The mixture is applied to a thoroughly pre-cleansed glass block and to a black, high gloss, lacquered glass block using a spreader with a 200 µm slit.

The test results in CC lacquer are given in table 5 and in DD lacquer in table 6. For comparison the precipitated silicas according to DE 38 15 670 and the commercial product NIPSIL E 1009 are also given. A comparison of the data determined can be obtained from the tables.

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Table 1

Ex.	Speed of mill	Speed of classifier	Class-ifier air	Throu-ghput	Particle size (Malvern)				Grindo	Gloss		Sheen	Roughness		Viscosity	Thickness of coating
	rpm	rpm	m^3/h	kg/h	d 4.3	d 10	d 50	d 90	µm	60°	85°		RZD	Ra	s	µm
1 a	10700	11000	175	10	8.34	4.48	7.03	12.89	23	23.8	72.0	48.2	2.27	0.27	36	30
1 b	10000	10500	180	15	9.76	4.53	7.11	15.64	27	21.8	70.3	48.5	2.37	0.28	36	30
1 c	10000	9000	200	30	9.34	4.52	8.03	13.67	28	24.7	67.9	43.2			34	28
1 d	10000	10000	145	15	9.97	4.27	6.78	18.13	33	26.0	73.4	47.4			38	29

Table 2

Ex.	Speed of classifier	Milling air	Throu-ghput	Particle size (Malvern) micrometers (µm)				Grindo	Gloss		Sheen	Roughness		Viscosity	Thickness of coating
	rpm	m^3/h	kg/h	d 4.3	d 10	d 50	d 90	µm	60°	85°		RZD	Ra	s	µm
2 a	11000	150	20	6.49	3.74	5.95	9.7	23	16.6	66.4	49.8	2.24	0.28	36	40
2 b	11000	150	40	12.9	3.69	6.68	24.3	23	21.9	58.0	36.1	2.00	0.24	39	39
2 c	10000	150	20	11.5	4.98	8.47	17.9	27	16.6	58.8	42.2	3.24	0.42		
2 d	8000	150	30	12.2	5.76	11.5	19.5	39	15.6	43.8	28.2	4.90	0.55	36	42
2 e	11000	150	30	7.6	3.55	6.1	12.44	24	21.1	56.4	34.3				

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Table 3

Classifying precipitated silica, prepared according to example 1c

Ex.	Fraction	Speed	Class-ifier air	Throu-ghput	Particle size (Malvern)				Grindo	Gloss		Sheen	Roughness		Viscosity	Thickness of coating
		rpm	m^3/h	kg/h	d 4.3	d 10	d 50	d 90	µm	60°	85°		RZD	Ra	s	µm
3 a	fine	16000	53	4.3	7.42	4.24	6.78	11.13	22	25.3	75.7	50.4			23	30
	coarse				12.07	8.05	11.28	16.98	33	12.1	27.6	15.5			21	30
3 b	fine	16000	66	2.0	6.84	3.95	6.30	10.11	23	26.2	74.9	48.7			23	30
	coarse				11.18	8.28	10.83	14.46	33	12.3	28.4	14.1			21	30
3 c	fine	13000	117	6.0	7.42	4.24	6.82	11.07	22	23.1	71.9	48.8	2.13	0.26	23	30
	coarse				11.08	8.03	10.73	14.48	33	13.9	35.6	21.7			21	30

Classifying precipitated silica, prepared according to example 2c

Ex.	Fract-ion	Yield	Speed of classifier	Milling air	Throu-ghput	Particle size (Malvern)				Grindo	Gloss		Sheen	Roughness		Viscosity
		%	rpm	m^3/h	kg/h	d 4.3	d 10	d 50	d 90	µm	60°	85°		RZD	Ra	s
4 a	fine	85	13000		2.1	6.84	3.85	6.26	10.10	29	19.8	70.3	50.7	2.2	0.27	28
	coarse	15				10.17	8.32	9.91	12.35	29	10.9	31.2	20.3			24
4 b	fine	66	16000		2.1	7.37	3.01	4.84	11.08	17	21.8	77.6	55.8			28
	coarse	34				9.26	8.45	9.28	10.4	27	10.5	38.2	25.7			24

Table 4

	Particle size (um)				Grindo	Gloss		Sheen
	d 4.3	d 10	d 50	d 90	um	60°	85°	
Comparison example 4	18.7	6.4	14.9	35.1	> 100	10.5	15.2	4.7
Comparison example 5	12.8	3.4	6.2	20.7	85 Specks, air bubbles	18.4	52.4	44.0

Table 5

CC lacquer

Example according to:	DE 38 15 670	1 a	1 b	2 c	NIPSIL B 1009
Flowtime in DIN seconds at 23 °C	140	149	148	135	118
Thickness of coating in um	23	22	24	23	23
60° reflectometer value (DIN 67530)	36.9	36.7	36.3	37.7	44.4
85° reflectometer value (DIN 67530)	79.3	78.9	77.7	77.5	86.5
Sheen	42.6	42.2	41.4	39.8	42.1

Table 6

CD lacquer

Example according to:	DE 38 15 670	1 a	1 b	1 c	NIPSIL E 1009
Amount of matting agent added	7.5	7.5	7.5	7.5	7.5
Flowtime in DIN seconds at 23 °C	31	42	41	32	23
60° reflectometer value (DIN 67530)	19.5	30	30.2	43.7	90.4
85° reflectometer value (DIN 67530)	55.6	68.1	58.2	74.9	97.5
Macbeth RD 918 densitometer value measured using yellow filter	2.12	2.31	2.17	2.16	2.3

Example 7

The matting efficiency is determined in a number of different lacquer systems, wherein the preparation and application of the lacquer took place under identical conditions each time.

A high matting efficiency means a low requirement (concentration) of matting agent in order to achieve a specific degree of gloss (measured at an angle of 60°C (sic)). The matting efficiency of unknown matting agents is determined in a relative manner, i.e. by comparison with known matting agents, so that variations in the determination of the degree of gloss (depending on the mode of preparation and application of the lacquer) are avoided. One important physico-chemical parameter which has a critical effect on the matting efficiency of silica is the particle size distribution of the silica. Basically, it has been shown that with identical precipitation processes the matting efficiency of the precipitated silica decreases with decreasing particle size (and vice versa). Fine fractions of precipitated silica have a lower matting efficiency than that of a more coarsely milled fraction.

The high matting efficiency of the precipitated silicas according to the invention is demonstrated as follows, in a variety of lacquer systems:

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Table 7: Test in alkyd/melamine lacquer

Lacquer system: alkyd melamine in accordance with formulation
Product from example 2c has higher matting efficiency than Syloid ED 5, although this product is more finely divided. Furthermore, product 2a is more efficient than Nipsil E 1009 and Syloid ED 3.

Product prepared according to example	Weight added	Particle size d4.3	Particle size d10	Particle size d50	Particle size d90	Grindometer	Gloss 60°	Gloss 85°	Sheen	RZD roughness (A/M)	Ra roughness (A/M)	Viscosity	Thickness of coating
	g		µm	µm	µm	µm						s	µm
1 + 3	4	12.32	6.58	11.48	18.83	32	16.0	43.0	27.0	3.48	0.46	34	32
1 + 3	4	11.85	5.99	10.90	18.70	34	16.0	46.0	30			37	32
2	4	12.22	5.76	11.63	19.50	40	16.4	45.0	28.6	4.30	0.55	38	42
OK 520	4			7.20		31	16.5	64.0	47.5	3.05	0.36	38	37
2	4	11.50	4.99	8.47	17.97	30	16.6	58.8	40.2	3.24	0.42	38	38
2	4	10.90	5.55	10.41	16.46	37	16.9	47.8	30.9			38	27
1	4	13.24	6.42	12.90	20.40	33	17.8	43.6	25.8			38	30
1 + 3	4	12.32	6.58	11.48	18.83	33	17.9	50.2	32.3	3.43	0.46	33	40
Syloid ED 5	4	10.47	6.30	9.56	16.62	32	18.7	51.0	32.8	3.65	0.46	32	41
1 + 3	4	8.85	4.50	8.37	13.19	25	19.8	61.9	42.1	2.80	0.35	37	32
1 + 3	4	8.85	4.50	8.37	13.19	25	21.0	63.0	42.0			34	40
1	4	11.37	5.81	10.95	17.12	34	21.5	55.2	33.7			35	28
1	4			7.10		27	21.8	70.3	48.5	2.37	0.28	35	
Syloid ED 3	4	6.04	3.62	5.54	8.88	21	22.0	73.0	51.0	2.03	0.24	35	34

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Product prepared according to example	Weight added	Particle size d4.3	Particle size d10	Particle size d50	Particle size d90	Grindometer	Gloss 60°	Gloss 85°	Sheen	RZD roughness (A/M)	RZD roughness (A/M)	Viscosity	Thickness of coating
	g		µm	µm	µm	µm						s	µm
Nipsil E 1009	4	7.92	4.34	6.97	12.51	27	22.0	70.0	48.0	2.44	0.28	38	32
OK 607	4	4.60		4.20		18	22.5	78.5	58.0	1.70	0.20	35	32
2 + 3	4	6.84	3.95	6.28	10.10	22	22.9	74.6	51.7	2.20	0.27	35	39
2	4	12.47	4.03	7.17	29.37	27	23.1	74.1	51.0	2.08	0.26	34	41
1	4	8.34	4.48	7.03	12.89	23	23.8	72.0	48.2	2.27	0.27	38	30
1	4	10.10	5.03	7.80	14.71	23	24.1	70.7	46.6			38	30
1	4	8.52	4.84	7.57	12.94	23	24.4	71.0	46.6			38	30
1	4	9.34	4.52	6.03	13.87	28	24.7	67.9	49.2			34	28
1 + 3	4	7.42	4.24	6.82	11.07	24	25.0	79.0	48.0	2.13	0.28	38	34

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Table 6: Tests in DD lacquer

Lacquer system: DD lacquer in accordance with formulation
Comparison example: Syloid ED 3

Product ref.	Weight added g	Malvern value d4.3 µm	Particle size d10 µm	Particle size d50 µm	Particle size d90 µm	Grindometer (A/M) µm	Densitometer value	Gloss 60°	Gloss 85°	Sheen	Roughness RZD (A/M)	Roughness Ra (A/M)	Viscosity s	Thickness of coating µm	Lacquer system
2b	7.65	12.93	3.69	6.68	24.35	25	2.11	25.0	66.2	41.2	2.00	0.24	n.m.	ca. 40	DD
2d	8.00	12.22	5.76	11.53	19.50	40	2.16	24.7	40.3	15.6	4.30	0.55	32	ca. 40	DD
3c	8.2	7.42	4.24	6.82	11.07	22	2.12	25.0	66.6	40.6	2.13	0.26	53	ca. 40	DD
2a	8.24	6.49	3.74	5.65	9.70	24	2.11	24.5	59.7	35.2	2.24	0.28	55	ca. 40	DD
1a	8.41	8.34	4.48	7.03	12.89	25	2.08	25.0	60.9	35.9	2.27	0.27	n.m.	ca. 40	DD
Precip silica	10.1	7.83	4.67	7.17	11.58	23	2.01	25.0	61.9	36.9	1.95	0.24	53	ca. 40	DD
Syloid ED 3	10.7	6.04	3.62	5.54	8.88	21	2.24	25.0	68.2	43.2	2.03	0.24	52	ca. 40	DD

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Table 9: Tests in DD lacquer

Lacquer system: DD lacquer in accordance with formulation
Comparison example: Nipsil E 1009

Product ref.	Weight added g	Particle size d4.3 µm	Particle size d10 µm	Particle size d50 µm	Particle size d90 µm	Grindometer value µm	Densitometer value	Gloss 60°	Gloss 85°	Sheen	Roughness RZD (A/M)	Roughness Ra (A/M)	Viscosity s	Thickness of layer µm	Lacquer system
2b	7.65	12.93	3.69	6.68	24.35	25	2.11	25.0	68.2	41.2	2.00	0.24	n.m.	ca. 40	DD
1a	8.41	8.34	4.48	7.03	12.69	25	2.08	25.0	60.9	35.9	2.27	0.27	n.m.	ca. 40	DD
Nipsil E 1009	11.3	7.92	4.34	6.97	12.51	27	1.96	25.0	60.5	35.5	2.44	0.28	35	ca. 40	DD

Table 10: Tests in coil coating lacquer

Lacquer system: coil coating lacquer in accordance with formulation

Product prepared according to example	Weight added	Particle size d4.3	Particle size d10	Particle size d50	Particle size d90	Grindometer	Gloss 60°	Gloss 85°	Sheen	Viscosity
	g	µm	µm	µm	µm	µm				s
HK 125	2.7		4.9	9.65	17.35	30	24.0	45.0	21.0	95
Syloid C 812	2		6.40	12.50	20.80	40	27.0	44.0	17.0	90
1	2	12.36	6.20	11.33	19.31	32	27.0	48.0	21.0	101
1	2	14.56	6.82	13.31	23.30	40	28.0	48.0	20.0	102
Lovel HSF	2		6.74	13.22	22.98	44	29.0	42.0	13.0	77

Table 11: Test in an acrylic dispersion (aqueous)

Lacquer system: acrylate dispersion (MB 2399-134), aqueous, from the Rohm and Haas company
Comparison product: AQ 75 N

Product name	Weight added g	Grindometer µm	Densitometer value	Gloss 60°	Gloss 85°	Sheen
TS 100 (Commercial product from Degussa AG)	0.25	41	2.5	69.3	92.3	23.0
TS 100 (Commercial product from Degussa AG)	0.5	41	2.4	56.1	87.0	30.9
TS 100 (Commercial product from Degussa AG)	0.75	41	2.28	44.7	82.0	37.3
TS 100 (Commercial product from Degussa AG)	1	41	2.17	30.4	73.4	43.0
Precipitated silica according to example 1b	1	29	2.09	31.3	53.8	22.5
AQ 75 N (Commercial product from Crosfield)	1	28	1.96	39.0	68.2	29.2
Precipitated silica according to example 1b	1.5	29	1.89	18.1	35.2	17.1
TS 100 (Commercial product from Degussa AG)	1.5	41	1.82	18.7	59.5	40.8
AQ 75 N (Commercial product from Crosfield)	1.5	28	1.91	31.9	61.0	29.1
Precipitated silica according to example 1b	2	29	1.79	12.4	26.2	12.8
TS 100 (Commercial product from Degussa AG)	2	41	1.8	15.3	66.0	50.7
AQ 75 N (Commercial product from Crosfield)	2	28	1.89	27.7	53.3	25.6
AQ 75 N (Commercial product from Crosfield)	2.5	28	1.87	21.3	51.5	30.2
AQ 75 N (Commercial product from Crosfield)	4	28	-	12.2	35.8	23.6

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Particle sizes are determined using a laser beam diffractometer from the Malvern company. Before the measurement, the silica is dispersed in water using a stirrer and ultrasound. This silica dispersion is then pumped round the instrument into the path of the beam (cell) using a pump.

Sheen is the difference in the degree of gloss measured at an angle of 85° and the degree of gloss measured at an angle of 60°.

The viscosity is determined using a 4 mm DIN cup. The flow time in seconds of the lacquer is measured in accordance with DIN 53 211.

Key to the abbreviations:

CC lacquer:	coil coating lacquer
DD lacquer:	Desmodur Desmophen lacquer Desmodur is a hardener based on isocyanates Desmophen is a polyalcohol, used as the binder component Desmodur/Desmophen are the registered trade names of Bayer AG
CAB	cellulose acetobutyrate
A/M	alkyd/melamine lacquer

Example 8

Coating with polyethylene wax emulsion.

Precipitated silica is prepared according to DE-OS 31 44 299, example 1. A wax emulsion (5 % wax with respect to silica) is added to the filter cake which has been liquefied under the action of shear forces (solids content 10.8 wt.%) and then stirred vigorously for a further 30 minutes. The wax emulsion is prepared in an autoclave which

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is steam-heatable and has a disperser. 4.8 parts by weight of an alkylpolyglycol ether (Marlowet® CFW) in 81.0 parts by weight of water at about 100°C is initially introduced. Then 14.2 parts by weight of low pressure polyethylene wax are added and heated to 130°C. On reaching 130°C, the disperser is switched on and dispersion takes place for 30 minutes. During this time the temperature is held at between 130°C and 140°C. After switching off the disperser and cooling to about 110°C, the final emulsion is discharged.

The polyethylene used is characterised by the following properties:

Average molecular weight	1000
Solidifying point	100 - 104 °C
Dropping point	110 - 117 °C
Density (g/cm^3)	0.93

The silica suspension coated with wax in this way is then dried in a rapid dryer (e.g. a spray drier) by atomising (e.g. two-fluid nozzle, 2.8 bar of atmospheric air). The dried product is milled in a mechanical classifier mill of the ZPS 50 type from the Alpine company. The physico-chemical data are given in table 12:

Table 12

		8a	8b
N_2 surface area	m^2/g	373	373
CTAB-surface area	m^2/g	333	333
DBP absorption	g/100 g	330	330
C content	%	3.4	3.4
pH		7.2	7.2
Compacted density	g/l	106	87
Particle size distribution (Malvern) in µm d_{90}		26.25	12.28
d_{50}		14.85	8.21
d_{10}		6.91	4.66

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Table 13: Alkyl melamine lacquer

			Comparison example *)	
	8 a	8 b	OK 500	OK 520
Flow time in DIN - seconds at 23 °C	31	29	30	32
Grindometer value µm	41	26	25	28
Thickness µm	30	29	29	28
60°-Reflectometer value (DIN 67530)	11.0	17.3	19.0	21.0
85°-Reflectometer value (DIN 67530)	24.3	42.9	69.5	76.9
Sheen	13.3	25.6	50.5	55.9

*) Degussa commercial product

Table 14: DD lacquer

			Comparison example *)	
	8 a	8 b	OK 500	OK 520
Flow time in DIN- seconds at 23 °C	23	27	29	30
Weight of matting agent added (g)	8.5	8.5	8.5	8.5
60°-Reflectometer value (DIN 67530)	21.6	34.4	69.9	8.6
85°-Reflectometer value (DIN 67530)	33.2	67.4	88.2	32.5
Sheen	11.6	33.0	18.3	23.9
Densitometer value - Macbeth RD 918 measured using yellow filter	2.12	2.32	2.31	1.69

*) Degussa commercial product

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Figure 1

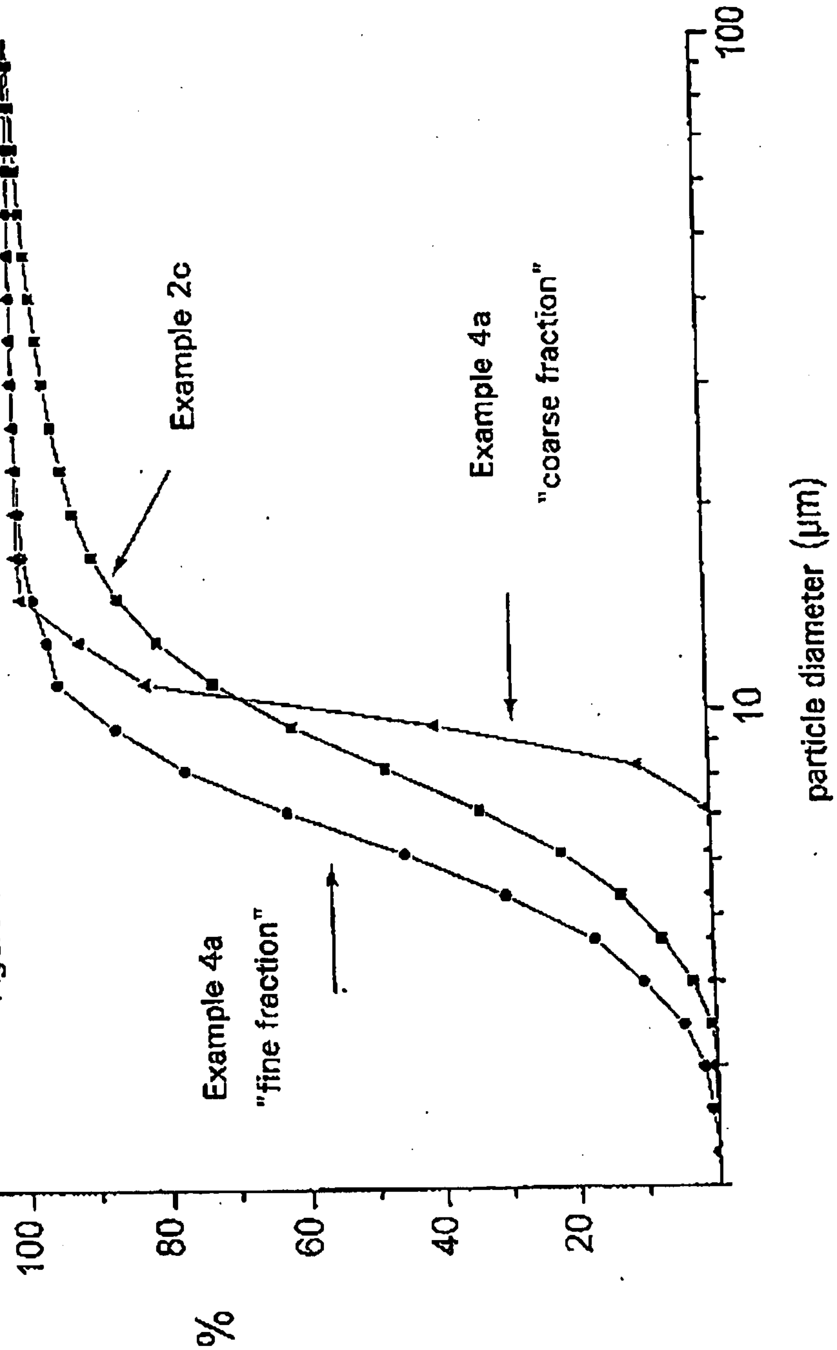

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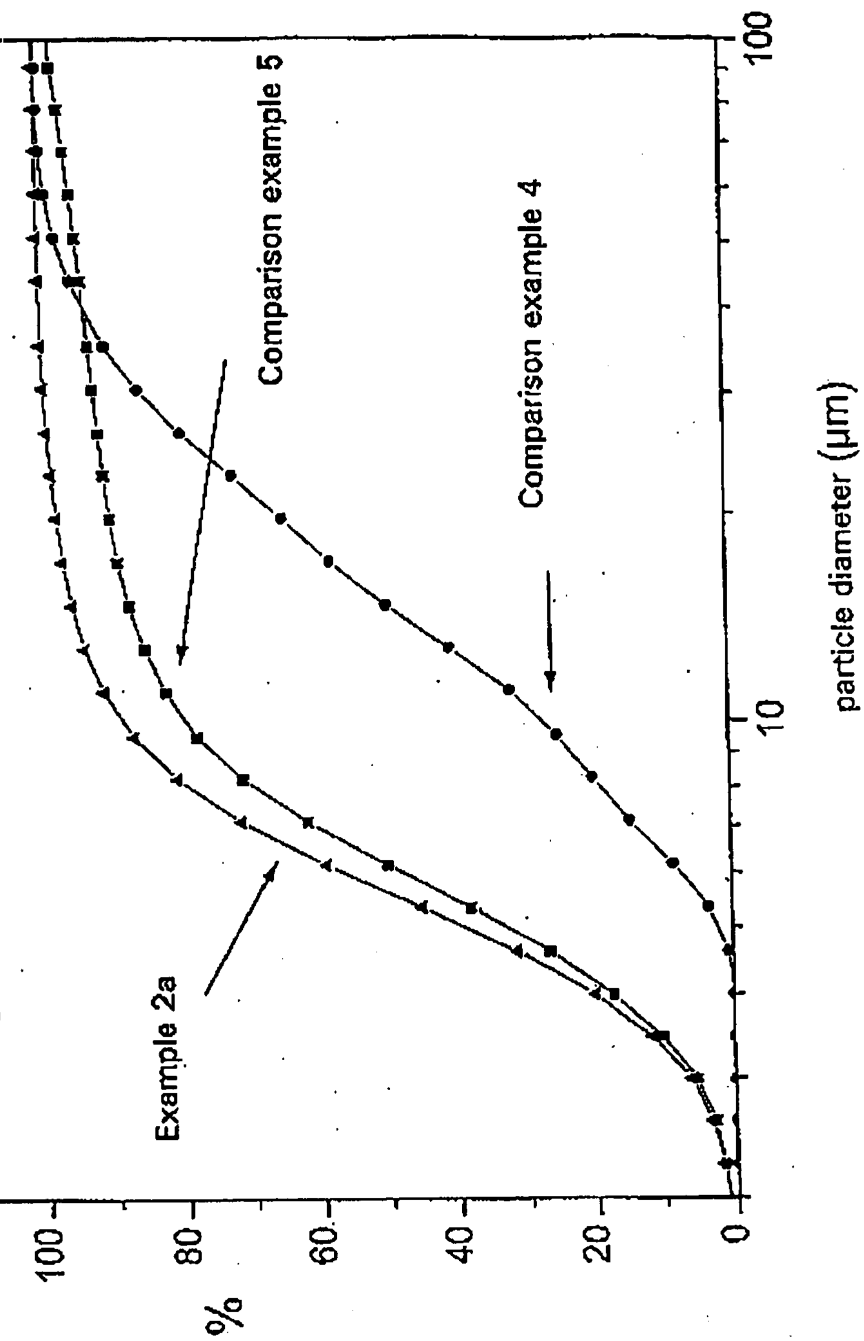
Figure 2



$$\frac{d_{90} - d_{10}}{2d_{50}}$$